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Original article

Identifying drivers of sewage-associated pollutants in pollinators across urban landscapes

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Abstract – Human sewage can introduce pollutants into food webs and threaten ecosystem integrity. Among the many sewage-associated pollutants, pharmaceuticals and personal care products (PPCPs) are consistent indicators of sewage in ecosystems and can also cause potent ecological consequences, even at minute concentrations (e.g., ng/L). Despite increased study over the past three decades, PPCPs in terrestrial ecosystems have been less studied than those in aquatic ecosystems. To evaluate PPCP prevalence and drivers in a terrestrial ecosystem, we analyzed managed and native bees collected from agroecosystems in Washington State (USA) for PPCPs. Caffeine, paraxanthine, cotinine, and acetaminophen were detected in all three evaluated taxa (*Bombus vosnesenskii*, *Agapostemon texanus*, and *Apis mellifera*), with *B. vosnesenskii* and *A. texanus* having a higher probability of PPCP detection relative to *A. mellifera*. The probability of PPCP presence in all three taxa increased in landscapes with more human development and greater plant abundance, with significant but negative interactions among these factors. These results suggest that human activity, availability of resources, and species-specific pollinator traits affect the introduction and mobilization of PPCPs in terrestrial ecosystems. Consequently, monitoring PPCPs and their ecological responses in terrestrial ecosystems creates opportunities to synthesize effects of sewage pollution across terrestrial and aquatic ecosystem types and organisms.

Arthropoda / emerging contaminants / entomology / natural history / persistent organic pollutants

1. INTRODUCTION

The introduction of wastewater and its byproducts into ecosystems can mobilize pollutants that reshape communities and food webs (Edmondson 1970). Historically, research on wastewater pollution has focused on changes in effluent nutrient concentrations as well as chemical and biological oxygen demand (Edmondson 1970; Brydon and Frodsham 2001; Tong et al. 2020). Recently, research emphasis has broadened to include micropollutants that are often found in sewage (Bernhardt et al. 2017). Pharmaceuticals and personal care products (PPCPs) in particular have garnered increased attention as an emerging organic micropollutant because they are globally pervasive and consistently associated with human

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sewage and pose potent, yet often uncertain, ecological consequences (Richmond et al. 2017; Meyer et al. 2019). Despite this increased attention, PCPPs tend to be less frequently sampled relative to macropollutants, which likely stem from their comparably high cost to collect and analyze (Meyer et al. 2019).

As PPCPs are consistently associated with sewage, their presence can indicate wastewater inputs into ecosystems. Previous surveys have shown that PPCPs, such as antibiotics, non-prescription and prescription drugs, hormones, and fragrances, are pervasive in subsurface and surface systems (Kolpin et al. 2002; Focazio et al. 2008; Wilkinson et al. 2022). Once introduced into ecosystems, PPCPs can propagate through food webs where they can be metabolized and accumulate within organisms (del Rey et al. 2011; Meador et al. 2016; Richmond et al. 2018). However, patterns of PPCP prevalence across taxa can be related to individual behavior and tissue allocation (Meador et al. 2016), and biological responses to PPCPs are often context-dependent, where responses can vary in terms of vulnerability of an ontogenetic stage at time of exposure and duration of exposure (Ginn et al. 2007). For example, activity and feeding rates of individual European perch increased when exposed to the anti-anxiolytic drug oxazepam, but sociality of perch populations decreased (Brodin et al. 2013). In insects, bumble bees fed nectar treated with caffeine increased foraging on nectars with similar aromatic compounds (Wright et al. 2013; Arnold et al. 2021). Overall, studies across diverse taxa suggest that biological responses to PPCPs are often deleterious, although specific responses may be uncertain (Richmond et al. 2017).

Although studies in aquatic systems suggest that PPCPs are ubiquitous and disrupt ecological processes, comparatively few studies have been conducted in terrestrial systems (Meyer et al. 2019). This imbalance in the literature creates opportunity to assess how PPCPs propagate through terrestrial food webs compared to aquatic food webs. For example, bees are terrestrial taxa that pollinate plants in natural and managed ecosystems (Kleijn et al. 2015; Iwasaki and Hogendoorn 2022; Papa et al. 2022). Bee pollinators may be commonly exposed to PPCPs through soil contact, interactions with plants, applications of biosolids for fertilizer, or through contamination of water in terrestrial ecosystems. For example, earthworms in soils experiencing biosolid application have been associated with increased PPCP concentrations (Carter et al. 2021). Due to the dramatic, recent declines in bee pollinators worldwide (Potts et al. 2010; Kleijn et al. 2015), bee taxa may be promising, societally important model organisms for expanding PPCP research from aquatic to terrestrial ecosystems.

Here, we assess PPCP prevalence and mobilization within pollinators in terrestrial ecosystems by focusing on three species: *Bombus vosnesenskii*, *Agapostemon texanus*, and *Apis mellifera.* Two of these bee species are wild (*B. vosnesenskii*, *A. texanus*), but *A. mellifera* is managed by humans. Our first goal was to identify species-specific patterns of PPCP presence. We predicted that taxa interacting with soil more frequently (i.e., ground-nesters; *B. vosnesenskii*, *A. texanus*) would be associated with a higher probability of PPCP presence than managed, colony-forming species (*A. mellifera*), as ground-nesting taxa would more likely encounter PPCPs within groundwater and biosolids. Our second goal was to assess potential drivers of PPCP presence. We predicted that PPCP presence would increase at sites with greater human development and sites with a higher density of floral plant resources, both of which may be sources of PPCPs (Carter et al. 2021; Meyer et al. 2022a). Overall, our study provides some of the first evidence for PPCP uptake in bee species, which have an exclusively terrestrial life cycle, while also demonstrating that landscape-context and species traits may affect exposure to PPCPs.

2. MATERIALS AND METHODS

2.1. Study system

Our study of PPCPs in bees was nested within a study of bee ecology on 36 small (< 25 ha), diversified farms and community gardens (e.g., farms and gardens with more than five unique flowering crops in simultaneous production) in western Washington (USA) (Bloom et al. 2022). Sites consisted of an assortment of private farms, farms owned by non-profit organizations, and urban gardens. For the present study, bees were collected from a subset of 10 locations that represented a range of human development (Figure 1). Each garden was at least 1 km apart for spatial independence (Bloom et al. 2022; Desaegher et al. 2022).

At each farm, we were not aware of direct sewage byproduct application both at the time of sampling as well as historically for each sampling location. Additionally, no sites were adjacently located to centralized forms of wastewater treatment, such as a wastewater treatment plant.

2.2. Bee and plant surveys

Bees were collected at each site using blue vane traps (SpringStar LLC, Woodinville, WA, USA), and bee bowls placed along a 50-m transect. All traps were filled with soapy water, where specimens could have remained up to 9 h. Upon trap collection, bee specimens were transferred to a 75% ethanol solution. After returning to the laboratory, all bees were rinsed with water, dried in a flask with kimwipes under low airflow, and then identified to species (Bloom et al. 2022). Of the 6539 specimens collected, we selected 101 for analysis, with 34 *B. vosnesenskii*, 32 *A. texanus*, and 35 *A. mellifera*; these taxa vary based on social behavior (social vs. solitary) and nesting strategies (cavity vs. ground-nesting). Because bees were collected across

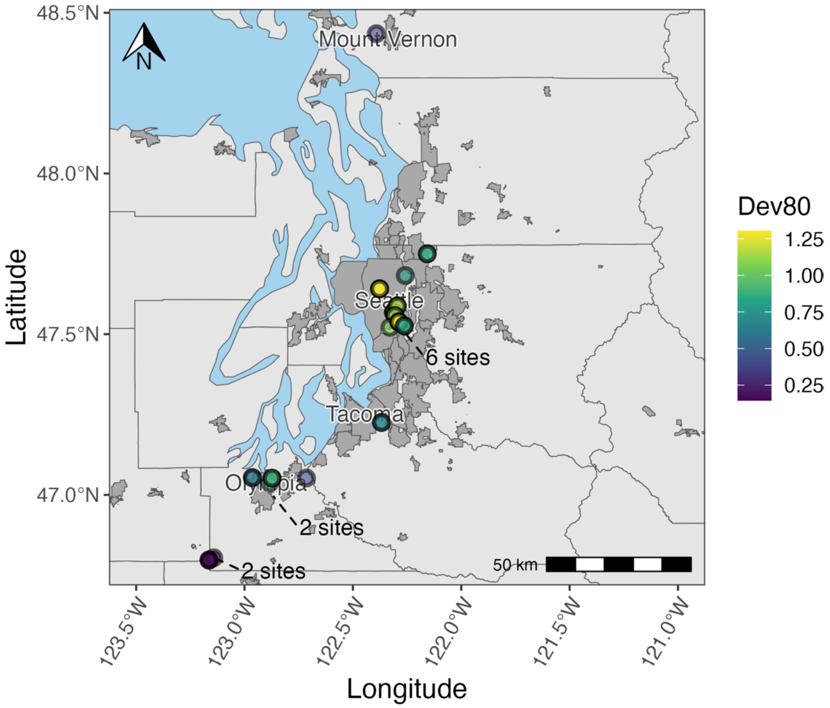

Figure 1. Map of bee sampling locations with sites colored by the value of 80% impervious development within 1 km of a sampling location. Gray polygons represent formal municipalities in the area. Dev80 values are arcsine-square root transformed for normality and consistency with modeling techniques. Data by OpenStreetMap (openstreetmap.org), under ODbL (openstreetmap.org/copyright). State data from US Census Bureau 2019.

3 years (2014, 2015, 2016) and three time points (spring, summer, fall), we used pinned specimens to have an approximately equal number of individuals across sampling periods. Specimen selection was stratified by species, year, time of collection, and sampling location, where each species-timing-year-site combination included at least two individuals. As we had more individuals of each species in any given time point, samples were haphazardly chosen from the larger collection within time points for each species to create a balanced set of samples. Only foraging taxa were selected, meaning that we were only able to assess bee PPCP conditions and not conditions that may be present in hives or nest sites. Additionally, as the selected individuals were taken from field surveys, we were not able a priori to isolate or cultivate individuals that were not exposed to PPCPs in these same sampling locations.

To assess the role of plant communities in affecting PPCP presence in bees, plant richness and abundance were measured at sites during sampling. Plants with flowers serving as resources for bee visitation (i.e., having pollen and nectar) were recorded along the same transect used for bee collection. All plants collected were in antithesis. A portable 1 × 1-m plot was placed over vegetation at 5-m intervals and all plants with open flowers were identified to species (Bloom et al. 2022). Each transect moved in a serpentine fashion across each study site. Summarized field variables are detailed in Table S1.

2.3. Landscape context

As sites were located along an urbanization gradient, we determined the proportion of developed landscape within 1 km of each site using the United States Department of Agriculture cropland data layer (CDL). Each pixel within the CDL classifies a 30 × 30-m area as one of 255 landscape classes. To characterize all human development in a landscape, we summed across low-, moderate-, and high-intensity pixels (Figure 1; Table S1) (Han et al. 2014).

2.4. PPCP extraction and quantification

Each dried bee sample was massed and then ground with a ceramic mortar and pestle. Bee PPCPs were extracted using a three-phase sequential extraction (Furlong et al. 2008; Brodin et al. 2013). First, 1.5 mL of methanol:water solution (7:3 ratio) with 0.1% formic acid was added into the mortar with the bee parts and transferred into a glass culture tube. Then, 1.5 mL methanol:water solution with 0.1% formic acid was used to rinse the mortar and pestle and was poured into the culture tube before centrifuging at 2000 rpm for 5 min. Following centrifugation, supernatant was moved to a clean culture tube and immediately sealed with parafilm. A second 1.5 mL of acetonitrile was next added to the first tube containing the bee tissue. The tube containing bee tissue was then vortexed and centrifuged twice. After each of the three extraction phases, the supernatant was placed under nitrogen flow in a 40 °C bath. Once samples were nearly evaporated, 1 mL of formate buffer was added to the sample, and the concentrated extract was transferred to a 1.5-mL amber glass autosampler vial. Samples were preserved in the dark at −20 °C until being analyzed with HPLC/MS.

2.5. HPLC/MS quantification

PPCP identification and quantification followed Furlong et al. (2008) and Brodin et al. (2013) with standards shown in Table S2. These methods targeted caffeine, acetaminophen/paracetamol, cotinine, codeine, warfarin, trimethoprim, sulfamethoxazole, diphenhydramine, thiabendazole, and paraxanthine/1,7-dimethylxanthine, as these PPCPs have been frequently monitored since the 1980s and would more likely be observed in samples relative to the over 4000 PPCPs on the global market (Daughton and Ternes

1999). HPLC eluents included a 10-mM formate buffer and 100% acetonitrile solution that varied in percent contribution over the quantification procedure (Table S3). All samples were analyzed using an Agilent 1260 Infinity II standard HPLC with a C18 Agilent column (2.1 × 150 mm). Sample injection volumes were 50 µL, and sample flow rate was 20 mL/min, as prescribed in Furlong et al. (2008). A main difference between our methods and others prior is that we split analyte signatures into separate channels to avoid peak interference (described in Table S2).

Standards were prepared in 70:30 methanol:water solution with 0.1 M formate buffer. Standards included concentrations of each analyte at 1 mg/L, 100 µg/L, 10 µg/L, 1 µg/L, 100 ng/L, 10 ng/L, and 1 ng/L. All standards were purchased from Sigma-Aldrich (St. Louis, MO, USA) and were rated at purities commensurate for HPLC analysis. All standards were prepared fresh on the day of HPLC/MS analysis. While Furlong et al. (2008) have a reported minimal detection limit of 5 ng/L, our methods were able to detect PPCP concentrations to 1 ng/L. To be conservative, we considered our minimal detection limit as 5 ng/L.

Samples were also analyzed in a way that accounted for potential cross-sample contamination and peak drift. Following standard samples, 2 blanks of 100% methanol were processed and assessed for contamination. Following the two blank samples, bee samples were processed in batches of 10 followed by 1 blank of 100% methanol, 1 standard (100 ng/L), and 1 blank of 100% methanol. This routine allowed us to purge the column from potential cross-sample contamination, to assess if samples were contaminating downstream samples within a batch, and to control for peak drift. No blanks contained PPCP residues, implying that cross-sample contamination was negligible.

For integrating peaks, personnel manually investigated each spectrum for each HPLC/MS channel and sample. If the personnel could clearly identify a peak, and the Agilent software was able to integrate a peak, the user manually performed all integrations. To ensure that signatures were consistent with potential peak drift, personnel manually compared the timing of each PPCP signature in the most recent standard analyzed with the samples preceding that standard.

Following peak quantification, we noticed that concentrations observed in bee samples tended to be bimodal, where PPCPs were either detected in higher concentrations or not detected entirely (Figure 2). We assumed the bimodality of these detections was likely a product of low-to-intermediate PPCP concentrations degrading since the time of specimen collection or diffusing from bee tissues as samples were originally collected in soapy water and then an ethanol solution before pinning (Bloom et al. 2022). To estimate bee PPCP presence, we reduced concentrations into categorical presence/absence informatics, implying that subsequent models and model interpretations should be conservative.

2.6. Quality control procedures

To account for potential contamination that might arise during the extraction and quantification process, laboratory personnel refrained from consuming caffeinated beverages, nicotine products, and non-prescription medications. Personnel wore an N95 mask and nitrile gloves to reduce chance of contamination during extraction.

During PPCP extraction, internal standards were not included. Consequently, we were not able to assess extraction recoveries. Therefore, concentrations reported may be lower than those reported, especially as lower concentrations tended to not be detected. To account for the lack of internal standards, we implemented a quadruple-blind sample processing routine to reduce probability of bias when analyzing samples. Throughout the entire specimen selection, PPCP extraction, peak quantification, and modeling procedures were performed by different personnel. Experimental autosampler vials were also haphazardly placed in the auto-sampler to

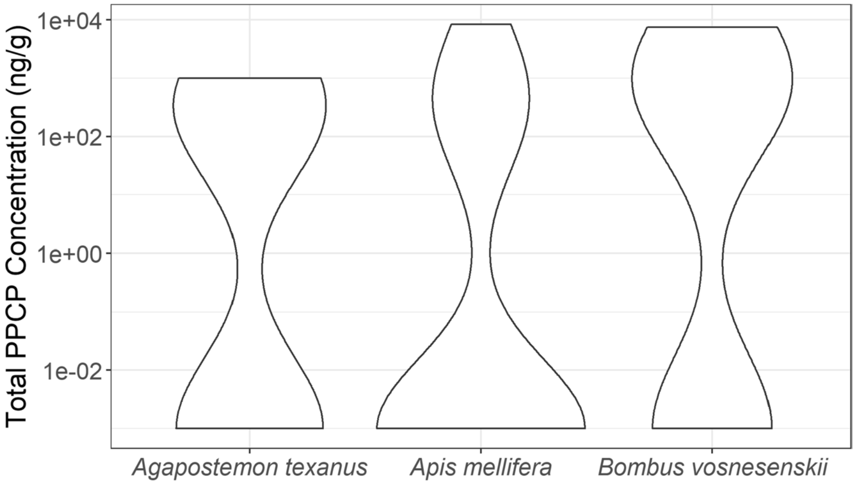

Figure 2. Violin plot of total PPCP concentrations (on log10 scale) in each of the three examined bee taxa (101 individuals in total; 34 *B. vosnesenskii*, 32 *A. texanus*, and 35 *A. mellifera*). Although the figure shows continuous PPCP concentrations, few samples contained "intermediate" PPCP concentrations, meaning that concentrations were either high (i.e., greater than 500 ng/g) or not detectable. Given that this framework seemed unrealistic and could be a product of how bees were preserved, our main analysis only focused on presence/absence of PPCPs in samples. Therefore, our logistic regression approach should be more conservative than continuous, linear analyses. Nevertheless, trends in PPCP concentrations mirror patterns observed in PPCP presence/absence, where *B. vosnesenkii* and *A. texanus* have higher concentrations and probabilities of PPCP presence relative to *A. mellifera.*

reduce likelihood of biases due to order in the auto-sampler. Personnel were therefore only aware of sample identity by the sample's unique numerical "Sample Identifier," which would not be intuitively related to an individual sample.

2.7. Modeling PPCP presence

To relate the probability of PPCP presence in bee tissues with potential environmental variables, we iterated through all possible combinations of logistic regression models, which used six predictor variables: bee and plant abundance, bee and plant diversity, percent development, bee taxon, and all two-way interactions between these variables. This technique is sometimes referred to as "exhaustive model selection" or "exhaustive feature searching" and has been employed in several biological contexts where non-linearities or the lack of multiscale data make process-based approaches unfeasible (Katz et al. 2015; Rheubert et al. 2020). To account for edge effects, all predictor variables were standardized by the perimeter:area ratio of their respective site. Once all possible models were generated, we selected the best model based on AICc, R^2, AUC, accuracy (percent true results), and p-value. When multiple models had similar performance (AICc within two points of lowest AICc value and R^2, AUC, and accuracy above the median of all models), best performing model coefficients were averaged to create a single statistical model.

To validate our results, we repeated analyses 1000 times with 80:20 train:test subsetted data, and compared the distribution of model parameters and pseudo-R^2 values from subsetted data to those models generated with the entire dataset. This subsetting routine allowed us to assess whether the distribution of possible model parameters was multi-modal and to evaluate whether the model constructed with 100% of the training data was overfit to those data (in the sense of Rheubert et al. 2020). In instances where model parameters generated with 100% of the training data represent the true environmental conditions that are predictive of PPCP presence in bee tissues, model parameters would come from unimodal distributions generated from models being trained by a subset of the data. Thus, if we compared the model parameters generated with 100% of the training data with the distributions of those generated with 80% of the training data, parameters fit with 100% of the training data should be representative of the general distributions.

All analyses were conducted within the R Statistical Environment (R Core Team 2022) using the packages glmulti (Calcagno 2019), tidyverse (Wickham et al. 2019), janitor (Firke 2020), MuMIn (Barton 2020), ggpubr (Kassambara 2019), ggeffects (Lüdecke 2018), lubridate (Grolemund and Wickham 2011), plotrix (Lemon 2006), Hmisc (Jr et al. 2020), corrplot (Wei and Simko 2017), ggrepel (Slowikowski 2019), ggspatial (Dunnington 2021), tigris (Walker 2021), cowplot (Wilke 2019), sf (Pebesma 2018), and readxl (Wickham and Bryan 2019).

3. RESULTS

3.1. Species-specific PPCP detections

We detected four PPCPs across all three bee species: caffeine, paraxanthine/1,7-dimethylxanthine, acetaminophen/paracetamol, and cotinine. We did not detect evidence of seven other PPCPs: codeine, warfarin, trimethoprim, sulfamethoxazole, diphenhydramine, thiabendazole, or albuterol. *Bombus vosnesenskii* (50% of samples; Tables S4 and S5) and *A. texanus* (44%; Tables S4 and S5) tended to have a higher odds ratio of PPCP presence relative to *A. mellifera* (26%; Tables S4 and S5).

3.2. Relating PPCP presence with human development and plant abundance

Across all bee taxa, the probability of PPCP presence increased in landscapes with greater human development and landscapes with higher plant abundance (Figure 3). However, there was also a significant negative interaction between human development and plant abundance. This negative interaction suggests the positive association between development and PPCP presence decreased in landscapes with greater plant abundance and that the positive effects of plant abundance on PPCP presence decreased in landscapes with greater development (Figure 3).

Our subsetting routine, which assessed the probability of observing the final model solely by chance, suggested that our final model coefficients were non-random (Figure 4). Model parameter estimates (Figure 4) as well as pseudo-R^2 values (Figure 5) were generally unimodal. Most subsetted model runs included human development and plant abundance as positive predictors of PPCP presence (Figures 3 and 4), and most coefficients for interactions between plant abundance and development were negative (Figures 3 and 4). Coefficients from the subsetting routine corresponded with patterns of PPCP detections for each taxon, where *B. vosnesenskii* had higher positive coefficients, and *A. mellifera* and *A. texanus* had lower negative coefficients (Figures 3 and 4).

4. DISCUSSION AND CONCLUSION

Our study demonstrates the presence of PPCPs in a terrestrial ecosystem and highlights that pinned insect specimens can be used along with environmental data to assess patterns of PPCP prevalence. These results are notable, considering that the study of PPCPs in terrestrial ecosystems is uncommon relative to aquatic environments (Meyer et al. 2019). Our results corroborate those results from aquatic systems, showing that PPCP presence can be influenced by human development and species-specific traits (Bendz et al. 2005; Meador et al. 2016) and that PPCPs can be mobilized in terrestrial food webs (Lagesson et al. 2016; Richmond et al. 2018).

Our results suggest that PPCPs are more frequently encountered in *B. vosnesenskii* and *A. texanus* than in *A. mellifera* (Figure 2; Table S4), implying that differences in species' life history traits may affect PPCP accumulation. Both *B.*

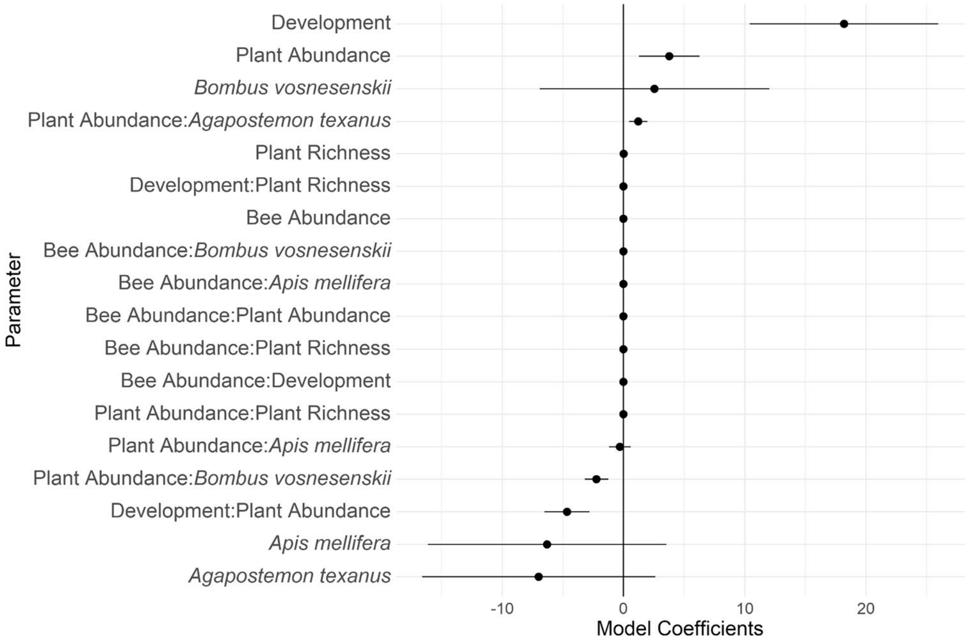

Figure 3. Coefficients for the final averaged model using all data for training. Points indicate the estimated model coefficient, and bars reflect the adjusted standard error. A value of 0 indicates that a variable does not discriminate between PPCP presence or absence. Values greater or less than 0 correspond to predictors that are more or less likely to be associated with PPCP presence, respectively. Parameters with a colon indicate two-way interaction terms. Parameters are arranged by decreasing coefficient estimate value.

vosnenskii and *A. texanus* create nests in the soil, which may increase exposure to human contaminants in the soil matrix and groundwater relative to *A. mellifera*, which is managed in artificial nests by humans (Gradish et al. 2019). Because bees were collected while foraging, our results are only pertinent to foraging bees and may not reflect conditions in hives or nest sites. These bees may encounter various point and non-point sources of PPCPs within their foraging radius. In particular, non-point sources commonly introduce PPCPs to aquatic and terrestrial ecosystems, and owing to heterogeneous, non-linear processes governing PPCP distribution in ecosystems, clearly associating PPCPs with their source can be challenging (Meyer et al. 2022a, b). As there was no direct evidence of sewage byproduct application and no adjacent wastewater treatment plants, all PPCPs detected were assumed to originate from non-point sources, such as groundwater transport from adjacent human developments.

Beyond sources of PPCP introduction, species' life histories and ecologies may also influence probability of PPCP uptake. From an ecological perspective, *A. mellifera* may occupy a distinct niche from the other taxa within the same location (Thompson and Hunt 1999; Leonhardt and Blüthgen 2012), where PPCP concentrations in certain food resources may generate variation in exposure. Niche partitioning of floral resources due to differences in traits, such as tongue length, could also mediate pollutant exposure and uptake (e.g., pesticides) (Brittain and Potts 2011). Further, age of specimen, ontogenetic shifts, and exposure time of resource use may explain some variation in PPCP presence (in the sense of Ginn et al. 2007). As bees were not assessed for age or sex prior to PPCP extraction, we are unable to assess how much variation

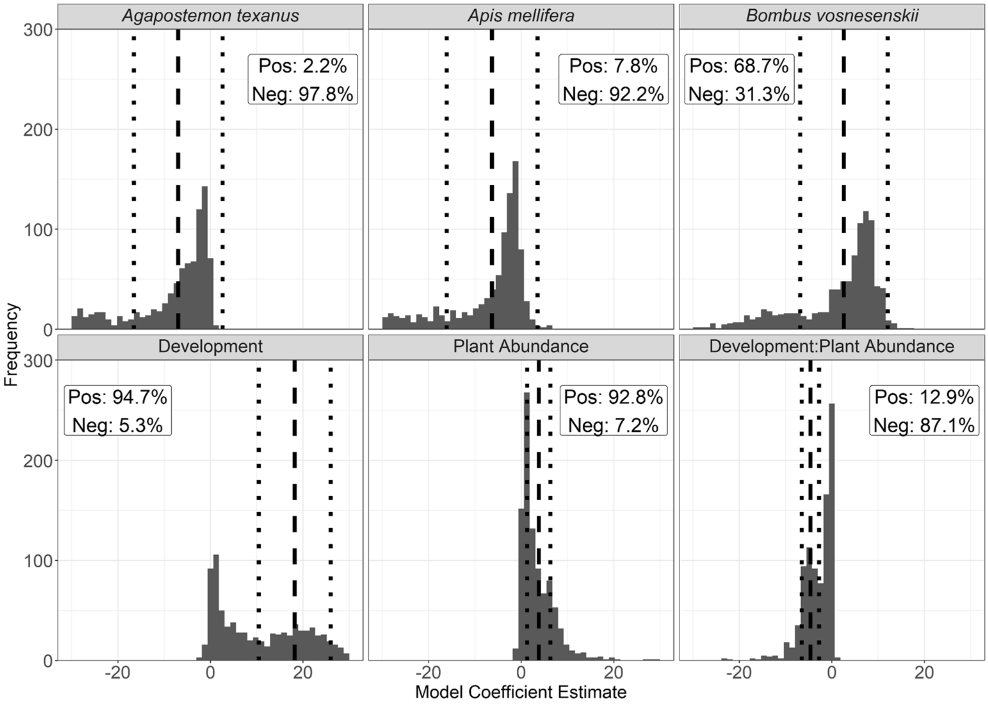

Figure 4. Distributions of most influential model coefficients for the final averaged model using 80% of original data as training data. Model averaging protocols were repeated 1000 times, with each iteration starting with a species-stratified, random subsample. Vertical, dashed lines indicate the coefficient of the parameter that appeared in the final averaged model, when 100% of the data were used. Vertical, dotted lines indicate the adjusted standard error of the coefficient of the parameter that appeared in the final averaged model, when 100% of the data were used. Labels within each facet reflect the percent of coefficients that were positive (i.e., greater than 0) and negative (i.e., less than 0).

could be explained by age, but given that patterns were observed across sites and years, our results suggest that inter-species variation in PPCP presence is greater than intra-species variation. In addition to pollinator traits, heightened PPCP exposure for *B. vosnenskii* and *A. texanus* may be due to production practices across sampling locations, where farmers may be applying wastewater byproducts to soil (Karnjanapiboonwong et al. 2010; Shahriar et al. 2021). While we are not aware of wastewater byproduct applications at our sampling locations (e.g., manure or sludge), prevalence of human-associated pollutants in conventional and organic farms is common (Humann-Guilleminot et al. 2019).

Beyond species-specific patterns, our results suggest that human development and plant abundance mediated PPCP presence across all three taxa. This result is consistent with previous findings in aquatic systems, where the source concentration (e.g., amount of development) and paths to trophic transfer (e.g., number of plants) are correlated with the presence of PPCPs at higher trophic levels (Richmond et al. 2018). For example, PPCP concentrations in aquatic systems can be directly proportional with human population size and inversely proportional to distance from a human population center (Bendz et al. 2005; Meyer et al. 2022b). PPCPs also can transfer between trophic levels (Richmond et al. 2018), which may lead to non-linear processes and consequences, such as our data showing a negative interaction between plant abundance and human development. Consequences of PPCPs within food webs are challenging to predict, although they can cause biological responses

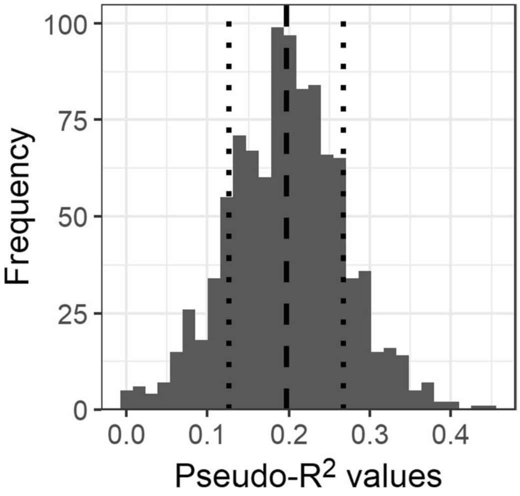

Figure 5. Distribution of pseudo-R^2 values from all averaged models in the permuted analysis. The vertical dashed line represents the mean of the distribution (pseudo-$R^2 = 0.197$), and vertical, dotted lines represent 1 standard deviation from the mean ($sd = 0.07$).

at physiological (Feijão et al. 2020), behavioral (Brodin et al. 2013), population (Hoppe et al. 2012), community (Lee et al. 2016), and ecosystem (Richmond et al. 2019) levels. Aside from PPCPs eliciting direct biological responses, PPCPs may co-occur with numerous human disturbances, such as nutrient pollution, that may further obfuscate clear associations between biological consequences and PPCP exposure.

To our knowledge, this is the first study to detect PPCPs in bee tissues that were preserved and pinned in a manner similar to natural history collections. While not originally intended for the present study, detailed field notes and extensive covariate collection created the opportunity for us to evaluate PPCP presence across taxa while testing potential mechanistic drivers of PPCP presence. Considering the growing number of pharmaceuticals (Daughton and Ternes 1999) and changes in pesticides used on national and global markets (Douglas et al. 2020), preserved collections, such as those in natural history collections, may offer ripe and previously untapped opportunities to explore contaminant accumulation and mixtures within biota. For example, lake sediment cores have been used to reconstruct inter-decadal PPCP contamination loadings (Anger et al. 2013). Similarly, natural history collections may empower reconstructions of exposure, accumulation, and co-contaminant histories, all of which may be useful for detailing how PPCP loading and mixtures may change through time.

4.1. Synthesizing PPCP patterns across ecosystem types

Over the past three decades, the study of PPCPs has expanded rapidly, and growing evidence suggests that PPCPs are pervasive micropollutants across aquatic and terrestrial systems alike. Our results suggest that much like aquatic systems, PPCPs tend to concentrate closer to areas with increased human development and have potential to enter food webs when vectors for trophic transfer are present. Additionally, our results demonstrate species-specific differences for PPCP uptake. Similar to patterns observed in PPCP accumulation in aquatic and riparian systems (Meador et al. 2016; Richmond et al. 2018), *B. vosnesenskii* and *A. texanus* had higher probabilities of PPCP detections relative to *A. mellifera*, implying that life histories, behavior, or physiological differences between taxa may play a role in PPCP uptake.

Broadly, our results present novel opportunities for assessing PPCP presence throughout food webs and suggest similarities to how terrestrial and aquatic systems accumulate PPCPs. Where PPCP data are rarer, pairing human development, environmental, and ecological data may aid managers in flagging systems that are more associated with PPCPs, and thus susceptible to declines in ecosystem function and services (e.g., pollination). Regardless of the exact trajectory, our study lays a foundation for future basic and applied PPCP research and creates a synoptic view of how organic contaminants may mobilize within aquatic and terrestrial environments.

SUPPLEMENTARY INFORMATION

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AUTHOR CONTRIBUTION

M. F. M., M. R. B., B. W. L., E. H. B., and D. W. C. designed the experiment. E. H. B. collected the field samples. M. F. M., B. W. L., and M. R. B. developed the script analyses and data visualization. M. F. M. and M. L. A. extracted the PPCPs and analyzed the HPLC/MS outputs. All authors contributed to text, edited, and approved the final manuscript.

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AVAILABILITY OF DATA AND MATERIALS

All data used to produce these analyses can be found in this project's companion Open Science Framework repository (Brousil et al. 2022).

CODE AVAILABILITY

All scripts used to produce these analyses can be found in this project's companion Open Science Framework repository (Brousil et al. 2022).

DECLARATIONS

Ethics approval This study did not require any ethics approval.

Consent to participate The manuscript had no human participants.

Consent for publication Data contributors acknowledged consent to use their data prior to submission of this manuscript.

Conflict of interest The authors declare no competing interests.

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